

**MICHIGAN-ONTARIO OZONE SOURCE EXPERIMENT
(MOOSE)**

Science Plan

May 25, 2021

EXECUTIVE SUMMARY

Southeast Michigan (SEMI) is currently designated as in Marginal Nonattainment of the U.S. federal ozone standard and is likely to be bumped up to Moderate Nonattainment based on monitoring data for the years 2018, 2019, and 2020. Many locations in southern Ontario also frequently exceed the Canadian ambient air quality standard for ozone. The Michigan Department of Environment, Great Lakes, and Energy (EGLE) seeks an attainment strategy for the SEMI ozone nonattainment area that remains open to all viable options as appropriate, including a U.S. Clean Air Act (CAA) Section 179B(b) international transport petition and demonstration, an exceptional event demonstration, or an ozone attainment plan and attainment demonstration. There is also interest from the Ontario Ministry of Environment, Conservation and Parks (MECP), Environment and Climate Change Canada (ECCC), and the U.S. Environmental Protection Agency (EPA) to better understand what contributes to elevated ozone levels in the Border region. To ensure a viable ozone attainment strategy, both in the short and long term, regulatory and scientific agencies, including EGLE, MECP, the U.S. EPA, ECCC, and other partners, have decided to conduct field studies in 2021 and 2022 to be known as the Michigan-Ontario Ozone Source Experiment (MOOSE).

MOOSE will consist of three sub-experiments with the following objectives:

Great Lakes Meteorology and Ozone Recirculation (GLAMOR)

- To understand and simulate complex 3D flows associated with lake breeze circulations;
- To understand and simulate the urban heat island (UHI) and its interaction with the lake breeze;
- To understand and simulate the impact of lake breezes and the UHI on ozone and ozone precursor transport;
- To understand and track the influence of urban emissions and land-lake breezes on urban oxidative capacity through nitrous acid (HONO) and related reactive nitrogen species.
- To determine the conceptual picture (mesoscale meteorological patterns and photochemical production locations) for ozone exceedances in the Border region;
- To select representative ozone episodes for each identified mesoscale pattern, which can then be used as model base case periods for future ozone attainment demonstrations; and
- To conduct modeling and data analyses in support of an ozone attainment demonstration or, if warranted, a CAA 179B(b) petition or ozone exceptional event demonstration.

Chemical Source Signatures (CHESS)

- To characterize the ozone precursor signatures at key monitoring stations in the Border region where design values are highest during ozone exceedances in a normal year;
- To characterize emission plumes from point sources, area sources, and major industrial sectors in the Border region and their impacts on ozone design values on both sides of the U.S.-Canada border;
- To develop emission source fingerprints for the most important industrial facilities and source sectors in the Border region;
- To characterize the horizontal variations (including upwind, interior, and downwind concentrations) of NO_x and VOC in SEMI;
- To perform receptor modeling, source apportionment, and ozone culpability analyses to improve emission inventories and inform potential control strategies; and
- To perform air quality model simulations of potential emission control strategies.

Methane Releases from Landfills and Gas Lines (MERLIN)

- To determine the natural gas leakage rate of pipeline or other infrastructure in SEMI;
- To quantify methane, formaldehyde, and other emissions from landfills in the Border region; and
- To determine the contributions of large methane sources to ozone exceedances in the Border region, thereby informing potential control strategies.

1. BACKGROUND

1.1 Air Quality Issues in Michigan, USA

1.1.1 Southeast Michigan Ozone Attainment Status

The U.S. National Ambient Air Quality Standard (NAAQS) for ozone is set at 70 parts per billion (ppb) by volume averaged over 8 hours. Attainment of the ozone NAAQS is based on a design value computed for each monitoring station in a regulatory monitoring network. The design value is defined as the three-year average of the yearly fourth highest daily maximum 8-hour average ozone concentration measured at a monitoring site. A design value exceeding 70 ppb at any monitoring site in a metropolitan area normally results in that area's being designated by the U.S. Environmental Protection Agency (EPA) as an ozone nonattainment area. The Southeast Michigan (SEMI) ozone nonattainment area consists of the seven counties of St. Clair, Macomb, Oakland, Livingston, Wayne, Washtenaw, and Monroe.

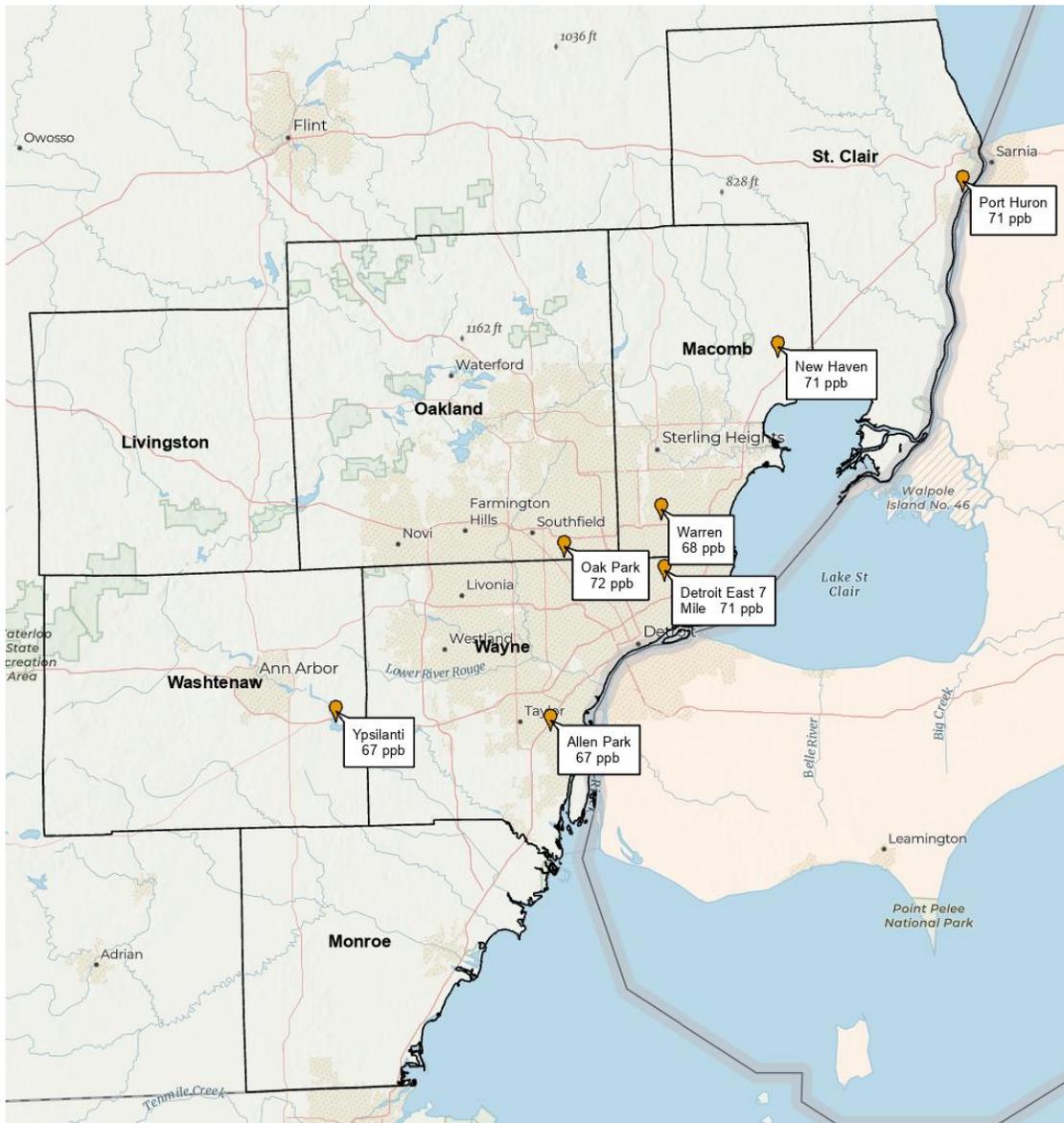


Figure 1. Monitoring sites and 2018-2020 ozone design values in SEMI nonattainment counties.

SEMI is currently designated as a Marginal Nonattainment area, the lowest nonattainment category, based on design values computed for the years 2015, 2016, and 2017. The next highest nonattainment category is Moderate Nonattainment, which imposes stricter requirements, including mandatory vehicle inspection and maintenance, higher industrial emission offsets (1.15-to-1), imposition of Reasonably Available Control Technology (RACT), and 15% Reasonable Further Progress (RFP) reductions in precursor emissions. A nonattainment area that fails to attain the ozone standard by the relevant deadline is normally “bumped up” to the next nonattainment category.

The attainment deadline for the SEMI region is August 3, 2021 based on design values computed for the years 2018, 2019, and 2020. Monitored ozone values for these three years (see Figure 1) indicate that SEMI will be bumped up to Moderate Nonattainment status, most likely around February 2022. This would require the State of Michigan to submit a State Implementation Plan (SIP) to the EPA that includes an ozone attainment plan and attainment demonstration, in which a computer model simulates the impact of control strategies intended to bring design values below 70 ppb.

1.1.2 Options for a Section 179B(b) Petition and Exceptional Event Demonstration

The SEMI non-attainment area is immediately across the Detroit River, Lake St. Clair, and the St. Clair River from two industrialized cities in Canada, namely Windsor and Sarnia. Ozone exceedances in SEMI normally occur with southwesterly winds, based on trajectory analyses performed by technical staff of the Lake Michigan Air Directors Consortium (LADCO). However, these same analyses indicate that SEMI ozone exceedances can sometimes occur during periods of easterly wind, when Canadian sources are likely to contribute to SEMI ozone design values. Regulatory relief from several of the nonattainment provisions of the U.S. Clean Air Act (CAA) may be obtained by filing a CAA Section 179B(b) petition that demonstrates attainment of the NAAQS “but for emissions emanating from outside the United States.” While this does not demonstrate actual attainment, it would allow SEMI to avoid the consequences of a pending or future bump-up if the petition is approved by the EPA.

In addition to International Transport petitions under CAA 179B(b), flagging monitoring data that have been impacted by exceptional events is also an option. A successful exceptional event demonstration can sufficiently lower ozone design values, and on that basis demonstrate attainment. An example of an exceptional event is a wildfire. For a wildfire exceptional event demonstration to be successful, the plume must be shown to impact the state on policy-relevant, high ozone days. The Department of Environment, Great Lakes, and Energy (EGLE) is currently pursuing wildfire exceptional event demonstrations for Michigan’s western nonattainment areas bordering Lake Michigan based on data from 2018-2020. After an initial analysis of relevant data, EGLE has not decided to pursue such a demonstration for the SEMI region for the same three-year period, while remaining open to the possibility in future years.

1.2 Air Quality Issues in Ontario, Canada

Air quality impacts all Canadians and affects many aspects of society, including human health, the natural environment, buildings and infrastructure, crop production, and the economy. Federal, provincial, and territorial governments in Canada share responsibility for air quality management. Under the Canadian Council of Ministers of the Environment (CCME), federal, provincial, and territorial governments work collaboratively to improve air quality by implementing the Air Quality Management System (AQMS)¹.

Ambient air quality in Canada is assessed in part by comparing measurements to the Canadian Ambient Air Quality Standards (CAAQS), which are health- and environmental-based air quality objectives to further protect human health and the environment and to provide the drivers for air quality improvement across the country. Currently, more than 25% of Canadians live in areas that exceed at least one of the 2020 CAAQS.

¹ Although Québec supports the general objectives of AQMS, it will not implement the System since it includes federal industrial emission requirements that duplicate Québec’s Regulation. However, Québec is collaborating with jurisdictions on developing other elements of the system, notably air zones and airsheds.

Overall, air quality in Ontario has improved over time as both ambient concentrations of common air pollutants and emissions to air have decreased over the last 10 years. Ozone and fine particulate matter, the main components of smog, remain as pollutants of concern. Some areas of Ontario continue to experience elevated levels of some pollutants due to local sources (e.g., industry, transportation), increasing background levels, and transboundary air pollution. Many locations in southern Ontario continue to exceed the ozone CAAQS. As more stringent ozone CAAQS come into force in 2020 (62 ppb) and 2025 (60 ppb), it may become even more difficult to achieve the standards. (Note that the statistical form of the CAAQS for ozone is identical to that of the U.S. ozone NAAQS.)

Smog-related air pollutants are generated both locally and regionally, and, with winds, can travel hundreds of kilometers, affecting areas far from the source of pollution. Long-range transport and transboundary flow of air pollutants play a significant role in Ontario's air quality. Typically, during the summer, elevated levels of these pollutants are associated with distinct weather patterns (e.g., slow-moving high-pressure systems originating from south of the lower Great Lakes) that result in the long-range transport of these pollutants into Ontario from neighboring U.S. industrial and urbanized states during south to southwesterly flow conditions. Transboundary sources from around the globe (global background) are also significant contributors to Ontario's ozone levels.

Ontario's framework for managing local and regional air quality issues has been developing for over 40 years. The framework has evolved from regulating industrial emissions from individual stacks, to protecting local air quality, and ultimately to addressing pollutants that have a regional impact, such as smog and acid rain. More recently, the Ontario Ministry of Environment, Conservation and Parks (MECP) has been looking at how to manage air issues that result from multiple sources of air pollution in a specific area, as well as how to deal with emissions that come from sources outside Ontario's boundaries.

Under AQMS the federal government has the responsibility to lead the actions and negotiations to address the transboundary flow of air pollutants originating from other countries with the involvement of affected provinces and territories. Canada has also been working closely with the United States under international agreements such as the Canada-U.S. Air Quality Agreement for many years. This collaborative project with Michigan, Ontario, the U.S. EPA, and other partners will further collective efforts to improve understanding of air quality in our common cross-border airshed.

1.3 The Need for New Observational Data

To control air pollution and avoid transboundary impacts in both Ontario and Michigan, an ozone strategy based on rigorous science is needed to support technical analyses and to pursue any of the available regulatory options. Although the regulatory systems in Canada and the U.S. are different, new observational data for the Border region, as a whole, will contribute to informing the responsible agencies as to which pollutants, sources, and sectors have the most important influence on air quality, and allow them to develop the most appropriate risk management actions. There are several scientific and technical issues that make this difficult to accomplish without additional research-grade measurement data.

1.3.1 Influence of the Lake Breeze and Urban Heat Island

Proximity to the Great Lakes poses difficulties in understanding how pollution is transported from land areas around the Great Lakes to the Border region. A fundamental need is to account for complex lake breeze effects. The previous 2017 Lake Michigan Ozone Study (LMOS) examined this issue in the context of the western Great Lakes region. A major conclusion of the LMOS was that very high resolution is required in a meteorological model (i.e., ~1 km horizontal grid cells) to be able to properly simulate lake breeze fronts and their effects on the transport of ozone and its precursors across Lake Michigan (LADCO, 2019). Likewise, the older 2007 Canadian Border Air Quality-Meteorology Study (BAQS-MET) demonstrated the importance of correctly simulating complex 3D air flows in modeling ozone over the eastern Great Lakes, as surrounding land areas may contribute up to ~30 ppb to regional background ozone (Makar et al., 2010; see Figures 2-4). The most recently available ozone model for SEMI only has 4 km horizontal resolution and is thus incapable of properly simulating lake breeze transport.

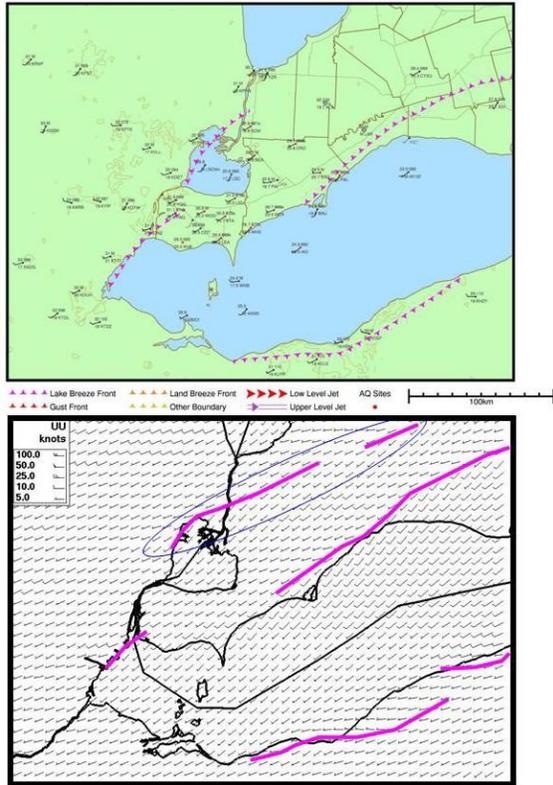


Figure 2. July 8, 2007, 1:00 p.m. local time. (a) meso-analysis lake-breeze front locations; (b) lake-breeze front locations inferred from convergence pattern of 2.5-km resolution model winds. (After Makar et al., 2010)

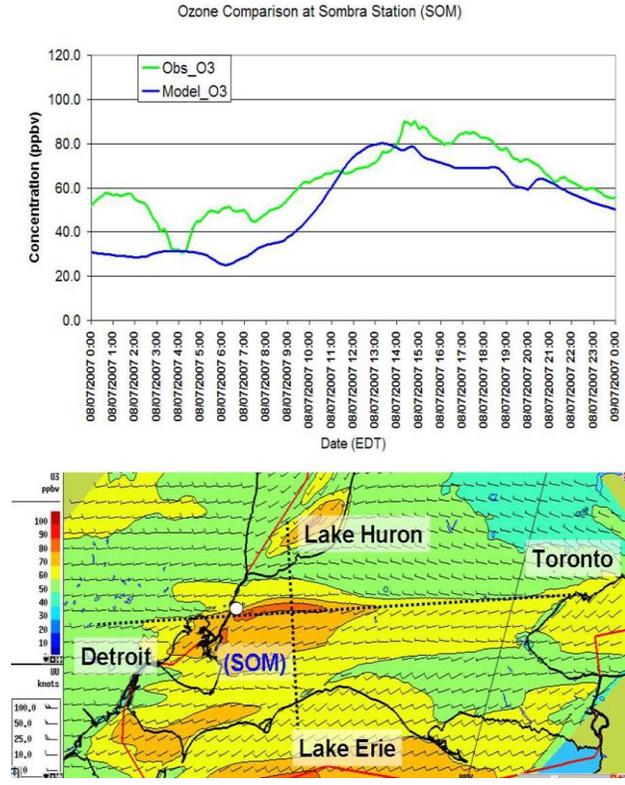


Figure 3. (a) Model-predicted ozone versus observations, July 8, 2007, Sombra (SOM) station. (b) Model-predicted ozone and winds at surface, 1:00 p.m. local time. (After Makar et al., 2010)

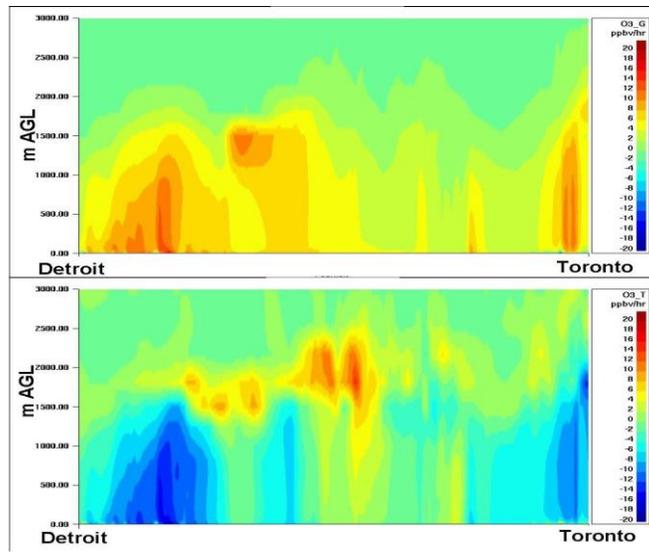


Figure 4. Model-predicted ozone mass tracking fields for July 8, 2007, 1:00 p.m. local time, Detroit to Toronto cross-section. (a) Gas-phase photochemical production, and loss; (b) total transport rate of change. (After Makar et al., 2010)

Another complicating factor in simulating ozone exceedances is the Urban Heat Island (UHI). The UHI influences meteorological parameters of importance to ozone photochemistry and transport, including surface air temperature, boundary layer height, and vertical mixing efficiency. In addition, the UHI can interact with the lake breeze, possibly intensifying some of its features due to increased updrafts associated with warmer surface temperatures.

The UHI was a prominent feature studied during the BAQS-MET campaign (Brook et al, 2013), and the 2015 Pan American Games in Toronto (Stroud et al., 2020), which have resulted in much higher resolution treatments of this phenomenon in Canadian meteorological and air quality models. These models now have horizontal resolutions as fine as 1 km. Stroud et al. (2020) discovered that a transition regime in ozone formation chemistry occurs in the updraft region of lake-breeze fronts. A chemical analysis along the trajectory of the lake-breeze circulation showed that in Toronto the most efficient ozone production occurs in the updraft region of the lake breeze front where the NO_x emissions are diluted.

A key need for ozone modeling in the Border region is more detailed meteorological measurements and higher resolution wind models to characterize 3D flow associated with lake breezes, and to account for the most important UHI influences on local and regional atmospheric chemistry and transport.

1.3.2 Chemical Fingerprints of Emission Sources

Recent weekday-weekend analyses funded by the Southeast Michigan Council of Governments (SEMCOG) indicate that the SEMI region is neither clearly VOC-limited nor NO_x-limited, but somewhere in between. A Photochemical Assessment Monitoring Station (PAMS), including an automated gas chromatograph (auto-GC) with flame ionization detection (FID) that measures speciated, hourly ambient concentrations of VOCs, has only recently been established at the East 7 Mile site in Detroit. As of this writing, it has yet to provide data to determine what emission sources to control to bring the SEMI region into ozone attainment. Chemical fingerprints identifying the dominant sources that contribute to ozone exceedances in the Border region would be very helpful in designing effective ozone control strategies.

Because of cleaner cars and other successful controls, urban VOC emissions have changed in recent years to favor oxygenated VOCs and other species associated with commercial and industrial Volatile Chemical Products (VCPs). VCPs may now make up more than half the mass and reactivity of urban VOC emissions (McDonald et al., 2018). While official VOC inventories are in the process of being adjusted in acknowledgement of this development (Seltzer et al., 2021), there is an ongoing need for new information and field measurements to constrain VOC emissions used as inputs to air quality models.

Among the most important oxygenated VOCs outside of VCPs is primary formaldehyde produced by incomplete combustion, as opposed to secondary formaldehyde produced by VOC reactions in air. Formaldehyde differs from most VOCs because it can efficiently generate an initial pool of atmospheric radicals that fuel ozone production. Unfortunately, formaldehyde emission inventories are unreliable, and measurements are needed to correct these inventories and avoid a deficit of ozone production in air quality models (see Figure 3a), including those used in attainment demonstrations (Olague et al., 2014).

Successful apportionment of emission sources may depend on clearly understanding concentration gradients of ozone precursors. An integrated strategy to characterize spatial gradients of NO_x and VOC has been successfully employed in other field studies, notably the 2017 Lake Michigan Ozone Study in the Chicago-Zion-Sheboygan area, and the 2018 Long Island Sound Ozone Study (LISTOS). These studies included airborne mapping measurements, such as from the Geostationary Trace gas and Aerosol Sensor Optimization (GeoTASO) instrument (Nowlan et al., 2016), in-situ vertical profiles of NO₂ and O₃, and ground-based column measurements of NO₂ and HCHO from Pandora spectrometers. GeoTASO and a sister instrument called the GEO-CAPE Airborne Simulator (GCAS) were developed as test beds for geostationary satellite instruments like NASA's Tropospheric Emissions: Monitoring of Pollution, TEMPO, instrument (Zoogman et al., 2017). GCAS/GeoTASO and Pandora were developed as validation instruments for OMI and TROPOMI measurements (Herman et al., 2009; Judd et al., 2020), and have been shown to provide valuable high time and spatial resolution measurements of NO₂ and HCHO columns. As in previous campaigns, remote sensing measurements can provide unique

perspectives on above-ground pollutant concentrations, regional transport, and diurnal variation, as well as the ability to learn how future measurements from TEMPO can aid in air quality analysis in the region.

Results of the MOOSE study will provide regional air planners with a better and more current understanding of ozone formation sensitivity to VOC and NO_x emissions in the Border region. Comparison to other urban areas where land/water interactions play a role in pollution transformation and transport (e.g., Lake Michigan, Chesapeake Bay, Long Island Sound) may help identify common or unique features among these regions that could be important in analyzing future satellite measurements. The nitrogen dioxide and formaldehyde measurements and high-resolution meteorological and chemical modeling will provide valuable information for developing more refined retrieval algorithms for TEMPO and companion missions. The Border region provides a robust test case for satellite observations due to its complex emissions profile (temporally and spatially) and the higher land/sea spatial resolution challenge within the relatively narrow and complex land/lake interface.

1.3.3 The Role of Methane Emissions

Methane is the second most important anthropogenic greenhouse gas after carbon dioxide, but it is also a global tropospheric ozone precursor. Because of its long atmospheric lifetime (~9 years), it has been thought of as too unreactive to be a significant local ozone precursor. However, methane emissions from natural gas distribution and end use may be 2-3 times larger than predicted by existing inventory methodologies and industry reports (McKain et al., 2015). Moreover, urban areas with corrosion-prone distribution lines may leak twenty-five times more methane than cities with more modern pipeline materials (von Fischer et al., 2017). Phillips et al. (2013) identified 3356 methane leaks in Boston with concentrations exceeding up to 15 times the global background level (1.8 ppm). Internal modeling experiments by the EPA Office of Research and Development showed that elevated methane levels in urban areas may increase local ozone levels by a few parts per billion (Dr. Rohit Mathur, personal communication), which is the typical width of ozone control strategies.

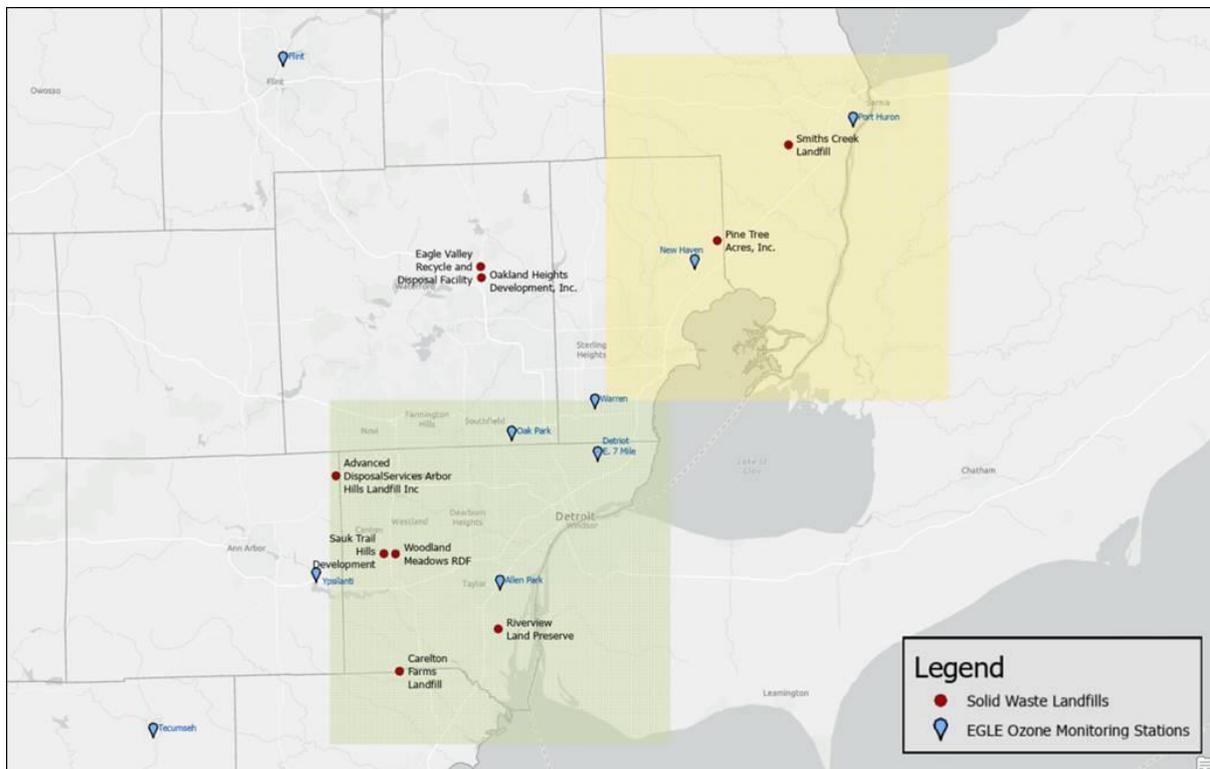


Figure 5. Locations of solid waste landfills and EGLE ozone monitoring stations in SEMI relative to proposed microscale modeling grids.

Another major source of methane is landfills. The California Methane Survey (Duren et al., 2019) deployed airborne visible and infrared imaging spectrometry to measure methane point sources (i.e., emissions from infrastructure elements or localized surface features) at 30 landfills and two composting facilities. These 32 sources collectively contributed about 43% to total point source emissions of methane in California, indicating the presence of super-emitters among the surveyed facilities. In the Great Lakes region, mobile infrared cavity ring-down spectrometry measurements by the EPA in 2016 and 2019 revealed ambient methane concentrations approximately 20 to 40 times the current global background level just outside one of the largest landfills in Michigan.

Besides methane, landfills are also a significant source of primary formaldehyde, mainly from landfill gas combustion in flares and in stationary engines at gas-to-energy conversion facilities. The combination of large emissions of methane and formaldehyde, along with combustion emissions of nitrogen oxides (NO_x) and VOCs other than formaldehyde (both in landfill gas and in products of incomplete combustion), may make landfills significant contributors to ozone. In SEMI, landfills are typically to the south and/or west (i.e., often upwind) of the key monitoring stations in the region (see Figure 5).

A key need for an ozone attainment demonstration is the quantification of the natural gas leakage rate in the Border region and of methane emissions from local landfills.

2. FIELD STUDY OVERVIEW

2.1 Organization

To ensure a viable ozone attainment strategy, both in the short and long term, regulatory and scientific agencies, including EGLE, MECP, the U.S. EPA, ECCC, and other partners, have decided to conduct field studies in 2021 and 2022 to be known as the Michigan-Ontario Ozone Source Experiment (MOOSE). Table 1 shows the main field study technical contacts for the various participating agencies.

Table 1. *Participating agencies and technical contacts*

Institution	Technical Contact	Position
Michigan Department of Environment, Great Lakes, and Energy (EGLE)	Dr. Eduardo (Jay) Olaguer Principal Investigator	Assistant Director, Air Quality Division
Environment and Climate Change Canada (ECCC) Environmental Protection Branch	Andrew Snider	Head, Project Management, Air Emissions Priorities Division
Environment and Climate Change Canada (ECCC) Science and Technology Branch	Dr. Craig Stroud	Research Scientist, Air Quality Research Division
	Dr. Felix R. Vogel	Research Scientist Climate Research Division
	Dr. Zen Mariani	Research Scientist Meteorological Research Division
	Katherine Hayden	Atmospheric Chemist, Air Quality Research Division
	Dr. Kevin Strawbridge	Research Scientist, Air Quality Research Division
Environment and Climate Change Canada (ECCC) Meteorological Service of Canada	Jacinthe Racine	Manager, Canadian Meteorological Centre Operations Division
Ontario Ministry of Environment, Conservation, and Parks (MECP)	Dr. Yushan Su	Senior Scientific Advisor, Air Monitoring and Modelling Section
	Dr. Rob Healy	Senior Scientist, Air Monitoring and Modelling Section
	Kelly Miki	Manager (Acting) Local Air Quality Permits
	Yvonne Hall	Supervisor, Air Modelling and Emissions Unit
Lake Michigan Air Directors Consortium (LADCO)	Zachary Adelman	Executive Director

U.S. Environmental Protection Agency (EPA) Region 5	Dr. Jennifer Liljegren	Physical Scientist
	Marta Fuoco	Physical Scientist
U.S. Environmental Protection Agency (EPA) Office of Research & Development Center for Environmental Measurement & Modeling	Dr. Rohit Mathur	Senior Scientist
	Dr. Lukas Valin	Research Scientist
U.S. Environmental Protection Agency (EPA) Office of Air Quality Planning & Standards	Dr. Kirk Baker	Physical Scientist
U.S. Forest Service (USFS), Northern Research Station	Dr. Joseph Charney	Research Meteorologist
National Aeronautical and Space Agency (NASA) Langley Research Center	Dr. Laura Judd	Associate Program Manager, Health and Air Quality Applied Sciences
NASA Goddard Space Flight Center	Dr. John Sullivan	Project Scientist, NASA Tropospheric Ozone Lidar Network
Aerodyne Research, Inc. (ARI)	Dr. Tara Yacovitch	Principal Scientist
University of Michigan (UM)	Dr. Stuart Batterman	Professor, School of Public Health
Brown University Institute at Brown for Environment & Society	Dr. Jiajue Chai	Assistant Professor (Research)
	Dr. Meredith Hastings	Professor, Department of Earth, Environment and Planetary Science
Wayne State University (WSU)	Dr. Yaoxian Huang	Assistant Professor Department of Civil and Environmental Engineering
State University of New York College of Environmental Science and Forestry (SUNY-ESF)	Dr. Huiting Mao	Professor, Associate Chair, Department of Chemistry
Colorado State University (CSU)	Dr. Joseph von Fischer	Professor, Department of Biology
Environmental Defense Fund (EDF)	Mary Gade	Advisor
Michigan Department of Technology, Management, and Budget (DTMB)	Shelley Jeltema	GIS Contractor

2.2 Period of Performance

Given the long timelines associated with ozone designations and emission control implementation, it is important to consider multiple years of data in understanding elevated ozone in the Border region, while minimizing the influence of non-conducive meteorology or other unusual circumstances such as the COVID19 pandemic. A concerted effort will be made to have as many of the project components and instruments operating simultaneously, to provide a robust description of elevated ozone conditions and precursor contributions. However, a multi-year effort provides flexibility for deployment of instruments that may not be available during certain periods, and also provides a longer time arc for understanding the impact of the unusual change in emissions activity related to the COVID19 pandemic.

MOOSE will have two phases: Phase I in 2021 and Phase II in 2022. Phase I will take place for six weeks in May and June of 2021. Phase II will occur during the summer of 2022. The measurement periods are intended to coincide with the most serious ozone exceedances. Historical data (see Table 2) indicate that late May, to early August are favorable times for ozone exceedances in the Border region. Available meteorological forecasts closer to the summer of 2022, as well as logistical considerations, will help to solidify the choice of study period for Phase II.

Table 2. Time periods corresponding to ozone and temperature metrics at the Detroit East 7 Mile site

Metric	2016	2017	2018	2019
Ozone 8hr 70+ppb	4/10 – 8/10	6/10 – 9/25	5/25 – 8/4	6/27 – 7/10
Ozone 1-hr. max	5/24 6/10 6/19 4/18 5/25	7/6 7/19 7/18 8/1 8/10	7/13 8/4 6/29 5/25 5/28	7/1 7/10 6/27 7/15 6/28
Max. 1-hour temp	7/22 7/23 8/10 8/12 6/11	6/12 6/11	5/28 6/17 6/18 5/5	6/27 6/29 5/25 6/26 6/27

2.3 MOOSE Sub-Experiments

Three main sub-experiments will occur during MOOSE, based on data needs identified in Section 1.2. The activities outlined below will proceed in 2021, while planning for a second phase of work in 2022 will be informed by lessons learned from 2021 activities, identified gaps and the availability of instrumentation that could not be deployed in 2021.

2.3.1 Great Lakes Meteorology and Ozone Recirculation (GLAMOR)

Performing Institutions: ECCC, MECP, EGLE, USFS, LADCO, WSU, SUNY-ESF, Brown University

Objectives:

- To understand and simulate complex 3D flows associated with lake breeze circulations;
- To understand and simulate the urban heat island (UHI) and its interaction with the lake breeze;
- To understand and simulate the impact of lake breezes and the UHI on ozone and ozone precursor transport;

- To understand and track the influence of urban emissions and land-lake breezes on urban oxidative capacity through nitrous acid (HONO) and related reactive nitrogen species.
- To determine the conceptual picture (mesoscale meteorological patterns and photochemical production locations) for ozone exceedances in the Border region;
- To select representative ozone episodes for each identified mesoscale pattern, which can then be used as model base case periods for future ozone attainment demonstrations; and
- To conduct modeling and data analyses in support of an ozone attainment demonstration or, if warranted, a CAA 179B(b) petition or ozone exceptional event demonstration.

Summary:

MECP will conduct enhanced monitoring at its Windsor West air monitoring station in Windsor, Ontario. MECP will deploy a meteorological sensor at 10 m above ground level to measure wind speed, wind direction, and temperature. Fast response measurements for NO, NO₂, SO₂, CO, O₃, black carbon, and PM_{2.5} will be performed. MECP will also deploy a PAMS GC instrument for hourly VOC monitoring and an Xact 625 instrument for hourly monitoring of trace elements. ECCC will co-locate a ceilometer to measure the height of the atmospheric boundary layer. Integrated 24-hour DNPH cartridge measurements of carbonyl species and 24-hour canister measurements of VOCs will also be conducted once every six days. A positive matrix factorization of all the hourly data at Windsor West will be performed to extract factors that can be interpreted along with meteorological back trajectories to provide insight into VOC source apportionment during the ozone exceedance periods. These receptor-based estimates can be compared with emission inventory-based estimates from numerical models, as discussed in Section 3.3.

Due to COVID19 restrictions, the deployment of additional instruments by ECCC at the ministry's Windsor West air monitoring station has been postponed to Phase II of MOOSE in 2022. These instruments include:

- either a SODAR or wind lidar to measure the vertical profile of wind in the boundary layer;
- an ozone lidar to measure the corresponding vertical profile of ozone;
- ozonesondes and a Vaisala system; and
- a Pandora instrument to measure column densities (and planar fluxes when paired with wind measurements) of ozone, nitrogen dioxide, and formaldehyde.

Measurements at the MECP's Windsor West air monitoring station will be complemented by additional meteorological and chemical measurements at the Detroit East 7 Mile PAMS station operated by EGLE. In addition to wind, temperature, and relative humidity measurements, EGLE will also operate a ceilometer to measure atmospheric boundary layer height and an auto-GC for VOC measurements. The East 7 Mile site also has instruments to measure NO₂ and O₃ concentrations.

An additional GLAMOR site will be operated by the USFS at the EGLE Port Huron monitoring station or other appropriate location in SEMI. Instruments will include a ceilometer and SODAR to perform continuous measurements of boundary layer height and wind profile.

Brown University, WSU, and SUNY-ESF will collaborate in performing field measurements of concentration and isotopic composition of NO_x ($\delta^{15}\text{N}$), HONO ($\delta^{15}\text{N}$, $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$), NO₂ ($\delta^{15}\text{N}$, $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$), HNO₃ ($\delta^{15}\text{N}$, $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$) and NO₃⁻(p) ($\delta^{15}\text{N}$, $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$). These measurements will: 1) constrain emissions, secondary production pathways, and sinks of HONO; 2) identify oxidation pathways of NO, NO₂, HONO, HNO₃ and NO₃⁻(p), and 3) quantify relative abundance of oxidants (O₃ vs RO₂). The field data will be used to develop an up-to-date and comprehensive chemical mechanism for reactive nitrogen species using a 0-D box model, and to add isotopic components to the EPA's National Emissions Inventory (NEI) concerning NO₂ and HONO. The new mechanism will then be applied to a 3-D chemical transport model (CMAQ), together with the improved EPA NEI, to quantify the role of HONO in urban to regional O₃ and secondary aerosol budgets.

After each phase of MOOSE, high-resolution meteorological simulations of the appropriate episodes will be performed using the Weather Research and Forecasting (WRF) and Global Environmental Multiscale

(GEM) models, with the aim of characterizing the dynamics of lake breezes, taking into account the influence of the local urban heat island. These meteorological simulations will then be used to drive air quality model simulations of the most interesting ozone episodes (see Section 3). Meteorological and air quality simulations will either be constrained (via 4D data assimilation) by or evaluated against the appropriate measurements at GLAMOR sites and other EGLE or MECP monitoring stations. The land surface scheme in the GEM model will be constrained with data from the Canadian Land Data Assimilation Surface (CALDAS) system, including lake water temperatures measured from buoys.

2.3.2 Chemical Source Signatures (CHESS)

Performing Institutions: ARI, UM, EGLE, ECCC, MECP, NASA

Objectives:

- To characterize the ozone precursor signatures at key monitoring stations in the Border region where design values are highest during ozone exceedances in a normal year;
- To characterize emission plumes from point sources, area sources, and major industrial sectors in the Border region and their impacts on ozone design values on both sides of the U.S.-Canada border;
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- To characterize the horizontal variations (including upwind, interior, and downwind concentrations) of NO_x and VOC in SEMI;
- To perform receptor modeling, source apportionment, and ozone culpability analyses to improve emission inventories and inform potential control strategies; and
- To perform air quality model simulations of potential emission control strategies.

Table 3. Chemical Instrument Manifest showing key instruments on board the mobile laboratories operated by ARI (AML) and UM (MPAL).

Measurement	LOD	Rate	Instrument	Platform
Select VOCs	30-300 ppt	1 s	Vocus proton transfer – time of flight mass spectrometer (Vocus PTR-ToF)	AML
Select VOCs	1-20 ppt	10 min	Gas chromatograph – electron impact – time of flight mass spectrometer (GC-EI-ToF)	AML
Methane (CH ₄), ethane, formaldehyde (HCHO), carbon monoxide (CO)	30 ppt - 3 ppb	1 s	Tunable infrared laser direct absorption spectrometer (TILDAS, multiple instruments)	AML
Carbon dioxide (CO ₂), CO, CH ₄ , H ₂ O, H ₂ S	1 ppb	1 s	Cavity ring-down spectrometers (Picarro G2401, Picarro G2204)	MPAL
Nitric oxide (NO) and Nitrogen dioxide (NO ₂)	0.3-1 ppb	1 s	Thermo 42i Chemiluminescence detector and Cavity Enhanced Phase Shift spectrometer; alternatively, TILDAS EcoPhysics CLD 700 AL	AML PAL
CO ₂	1.5 ppb	1 s	Licor 6262 non-dispersive infrared (NDIR) spectrometer	AML
Ozone (O ₃)	3 ppb	2 s	2BTech Ozone Monitor API 400A	AML MPAL
Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Pb, Al, Si, S, K, Ca	~1-10 ng/m ³	30 min	PM ₁₀ inlet, X-ray fluorescence, and β attenuation (Horiba PX375)	MPAL

Summary:

During CHESS, two mobile labs, operated by ARI and UM respectively, will be deployed in SEMI to measure a variety of chemical species (see Table 3). The mobile labs will measure source and air mass chemical fingerprints at various locations, guided by real-time meteorological and air quality forecasts provided by ECCC. Both mobile labs will be equipped with meteorological instruments, as well as a

Global Positioning System (GPS) to mark the precise locations of chemical and meteorological measurements.

The Aerodyne Mobile Laboratory (AML) will deploy a variety of advanced real-time sensors, including a Vocus Proton Transfer Reaction, Time-of-Flight, Mass Spectrometer for the measurement of a large suite of VOCs at high temporal (time response of 1 s) and mass resolution and very low limits of detection (<1 part per trillion). UM will deploy the Michigan Pollution Assessment Laboratory (MPAL) to measure sulfur dioxide (SO₂), carbon monoxide (CO), carbon dioxide (CO₂), methane (CH₄), hydrogen sulfide (H₂S), nitrogen oxides (NO_x, NO, NO₂), ozone (O₃), size-specific particulate matter (PM), black and brown carbon, and trace metals.

In Southern Ontario during Phase I, MECP will also deploy a mobile laboratory equipped with a GPS and real-time instrumentation, including a Proton Transfer Reaction Time-of-Flight Mass Spectrometer for the measurement of VOCs (time response of 1 s) and limits of detection <1 part per billion. The MECP mobile laboratory also features instrumentation for the measurement of ozone, sulphur dioxide, nitrogen oxides, aromatic VOCs (BTEX), and particulate matter at 5 s temporal resolution, as well as meteorological parameters. This platform will be used to measure air pollutant concentrations and chemical fingerprints immediately downwind of industrial sources in Sarnia and Windsor. It will also provide larger-scale spatial gradients of VOCs and ozone along the Ontario-Michigan border guided by ECCC meteorological forecasts. Data will be collected with 3 objectives: 1) to characterize the release of VOCs from point sources, 2) measure transboundary flow of pollution and 3) to map out the ozone spatial distribution for periods when 1-hr ozone exceedances are predicted in the study region.

To further understanding of the emissions and transport of key ozone precursors, as well as their spatial gradients, NASA will operate a Langley Research Center Gulfstream III (G-III) aircraft equipped with two instruments: the GeoCAPE Airborne Simulator (GCAS; Nowlan et al., 2016; Judd et al., 2020) and the Cloud Physics Lidar (CPL; McGill et al., 2002). GCAS is a UV-visible spectrometer that can measure below aircraft columns of NO₂ and formaldehyde at 350 x 350 m and 1 x 1 km spatial resolution, respectively, on the G-III. CPL is a backscatter lidar with three wavelengths that can provide profiles of clouds, aerosols, and smoke above and within the planetary boundary layer. During Phase I of MOOSE, NASA will perform at least 24 hours of instrumented flights over 3 days in June 2021 to reveal influence of emissions and meteorology on the structure of pollution plumes through repeated sampling over a region spanning from Monroe, Michigan to Sarnia, Ontario. This sampling strategy aims to simulate geostationary UV/VIS air quality mapping like those expected from NASA Tropospheric Emissions: Monitoring of Pollution Mission (TEMPO: <https://tempo.si.edu>).

After the field study, participants will collaborate in performing high-resolution receptor and inverse modeling to determine the contributions of various sources to ozone exceedances observed during MOOSE, as described in greater detail in Section 3.

2.3.3 Methane Releases from Landfills and Gas Lines (MERLIN)

Performing Institutions: UM, ARI, EGLE, EPA, ECCC, CSU, EDF

Objectives:

- To determine the natural gas leakage rate of pipeline or other infrastructure in SEMI;
- To quantify methane, formaldehyde, and other emissions from landfills in the Border region; and
- To determine the contributions of large methane sources to ozone exceedances in the Border region, thereby informing potential control strategies.

Summary:

MERLIN will occur during Phase I of MOOSE. UM, EPA, and CSU will each deploy GPS-equipped mobile laboratories with a Picarro cavity ring-down analyzer for methane and (in the case of EPA) formaldehyde, as well as supplementary instruments for measurement of combustion trace gases (in the UM and EPA mobile labs) and meteorological parameters. In addition, EGLE will deploy drone-mounted meteorological and chemical sensors to quantify emissions of methane, formaldehyde, and other ozone precursors from

selected landfills in SEMI. The ARI mobile lab may also be deployed during MERLIN to investigate emissions from natural gas pipelines and landfills, as well as the spatial, temporal, and chemical structure of any accompanying ozone plumes.

EPA Region 5 has developed a Geospatial Monitoring of Air Pollution (GMAP) platform with EPA's Office of Research and Development (ORD). GMAP implements an advanced technology that utilizes fast response instruments and a precise GPS that maps air pollution patterns around sources. This system uses a mobile platform to measure hydrogen sulfide (H_2S), methane (CH_4), benzene (C_6H_6), toluene (C_7H_8), ethylbenzene (C_8H_{10}), m-o-p xylene (C_8H_{10}), and ozone (O_3), along with meteorological parameters (wind speed, wind direction). By integrating these parameters with a concurrently collected geospatial tag from an incorporated GPS, the platform can be used to obtain highly sensitive ambient measurements to quantify air pollution concentrations, identify sources, and evaluate geospatial impact.

ECCC is hoping to conduct mobile methane surveys, as well as a landfill campaign, in southwest Ontario later in the summer or early fall of 2021. There are also plans to deploy the EM27/SUN solar tracking FTIRs for direct sun greenhouse gas measurements on a landfill. All these plans depend on COVID19 restrictions.

After the experiment, project partners will collaborate in performing data analysis and inverse modeling to quantify emissions of methane and any accompanying combustion tracers (in the case of landfills), as well their contributions to observed ozone exceedances.

3. MODELING, DATA ANALYSIS, AND INFORMATION MANAGEMENT

3.1 Information Management

EGLE is in the process of setting up a data management platform for MOOSE based on Geographical Information System (GIS) software (ESRI ArcGIS Pro). The platform will enable various layers of information, including those pertaining to EGGLE monitoring stations and MOOSE field study sites, industrial facilities in the SEMI area, and emissions inventories, to be (at least in some cases) interactively displayed over the Web and analyzed in systematic fashion. LADCO and EGGLE are also collaborating in the set-up of an emissions management platform for MOOSE based on the U.S. EPA's Emissions Modeling Framework (EMF).

An example of a GIS layer that EGGLE is currently building consists of underground pipeline segment shapefiles that will be useful in interpreting methane measurements made during the MERLIN sub-experiment in 2021. Access to proprietary and confidential business information will be limited on a strict need-to-know basis, and according to the appropriate guidelines and agreements between EGGLE and concerned parties.

EGGLE will also explore the possibility of providing a real time data broadcast capability during MOOSE, at least for the MERLIN sub-experiment. For example, UM mobile lab measurements may be made visible to field study participants over the Internet every few seconds so that the MPAL can serve as a "scout" to direct other mobile labs to high value measurement targets, and to perform coordinated upwind-downwind studies.

For long-term archival of MOOSE data, NASA will maintain a data repository, with submitted files subject to the ICARTT formatting convention. This file format had its origin in the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) field study in 2004, and has since been extensively used in other experimental campaigns. The ICARTT format is a text-based, self-describing, and relatively simple-to-use file structure composed of two sections: a header section (metadata) and a data section. The header section has the instructions for extracting data from the file and the critical information describing the data (e.g., data source, contact information, brief description of measurement technique, measurement uncertainties, and data revision comments) so that a user would have sufficient information to either make direct use of the data or contact the measurement PI to get further clarification on certain issues. The data section can accommodate different types of data, with an emphasis on standard time-series types of data, which is typical for in-situ chemical measurements. ICARTT is designed to fulfill the data management needs for all phases of a field study, i.e., field deployment, post deployment data processing and analysis, and publications.

3.2 Data Analysis

EGGLE will collaborate with field study partners in deploying data analytics to process and understand MOOSE field measurements. For example, EGGLE is working with CSU to deploy a Python-coded analysis tool for estimating natural gas pipeline leak volumes from real-time measurements of ambient methane concentrations. EGGLE is also currently working with UM to develop consistent quality assurance procedures for mobile measurements during MOOSE, and to perform hot-spot analyses of mobile lab data for methane using various mathematical techniques.

ESRI ArcGIS Pro has native data and statistical analysis tools. It also allows integration of Python and R scripts along with other applications to provide a workflow process that is well documented, consistent, and easy to use. Standard data formats will be employed whenever possible. A data dictionary will document data fields, calculation variables, and constants used in scripts and workflow processes. Instrument specifications assumed in measurement data analyses will also be documented.

Among the key data analysis efforts that will take place in the aftermath of MOOSE is receptor modeling by EGGLE and other MOOSE participants. For example, Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF) will be used to interpret chemical fingerprint data derived from the CHESS

sub-experiment. This will be enhanced by forward and inverse air quality modeling described below. The analysis and interpretation of measurements by factor and with high resolution air mass back trajectories will identify sources and the precursor VOC mixtures involved in the ozone production. For example, ranking VOCs by their OH-reactivity for the unique factors will be performed and compared to air quality model results for the same identified sources in model output.

Fast response VOC data can also be analyzed in terms of their diurnal pattern. Emissions, mixing, transport, and ozone photochemistry play different roles at different times of the day. In the early morning, stable surface conditions and rush-hour traffic emissions play a key role. At mid-morning, downward vertical mixing of regionally representative air occurs. In late morning and afternoon, photochemistry, mixing, and transport play a dominant role. Evaluating the model at these different times can help to understand what processes are responsible for biases.

3.3 Modeling and Forecasting

MOOSE presents an opportunity to explore meteorological and air quality modeling on finer scales than is the custom in SIP ozone attainment demonstrations. The necessity of this is conveyed by Figure 6, which shows tropospheric vertical columns of nitrogen dioxide at 250 m horizontal resolution in Chicago as measured by NASA's airborne GeoTASO UV/visible spectrometer in June, 2017 (Judd et al., 2019). Note the very fine horizontal filaments of NO_2 captured by the GeoTASO instrument. [*GeoTASO and GCAS have nearly identical capabilities for mapping NO_2 and HCHO .*] Datasets collected over the MOOSE domain from GCAS can help evaluate models at resolutions spanning from less than a kilometer up to the size of the domain sampled.

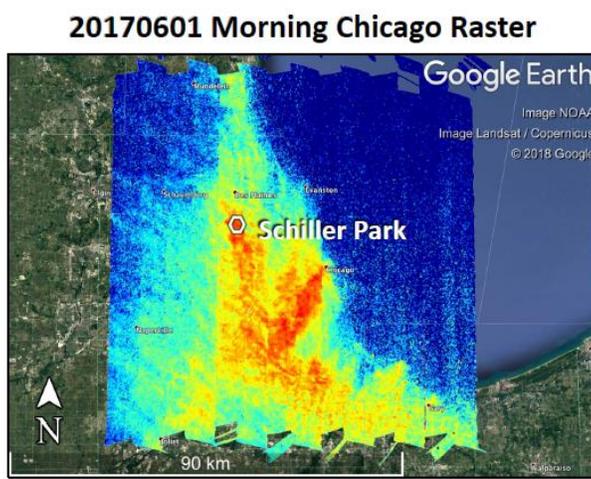


Figure 6. GeoTASO high resolution NO_2 Tropospheric Vertical Column (TropVC) measurements in Chicago as performed by NASA in June 2017 (Judd et al., 2019)

Olague (2012a,b) used a 3D Eulerian microscale chemical transport model at 200 m horizontal resolution to demonstrate that very fine ozone plumes may result from VOC and NO_x emitted by upstream and downstream petrochemical facilities, especially when accompanied by emissions of primary formaldehyde. Figure 7 shows fine-scale ozone and formaldehyde plumes from a large olefin flare event in the Houston Ship Channel as simulated by Olague (2012b). Olague (2013) and Olague et al. (2013) used an adjoint version of the model of Olague (2012a,b) to infer significant emissions of chemically reactive formaldehyde from petrochemical facilities based on research-grade field measurements.

Various models will be used by field study partners to analyze information during and after the MOOSE campaign. During the MOOSE intensive, ECCC will conduct high-resolution, real-time meteorological and air quality forecasts using the GEM-MACH model to guide the placement of the ARI mobile laboratory

during the CHESSE sub-experiment. This may be complemented by WRF meteorological forecasts by the USFS at 500 m horizontal resolution. LADCO will deploy the WRF meteorological and CAMx regional air quality models at 1.3 km horizontal resolution in the innermost grid to perform simulations of key wind and ozone episodes identified during MOOSE.

CAMx regional simulations will be complemented by even finer scale forward and inverse modeling by EGLE using a microscale chemical transport model at 200-400 m horizontal resolution with an intra-urban chemical mechanism valid for ambient NO concentrations above ~0.25 ppb. This mechanism will be more condensed and computationally efficient than a regional atmospheric chemical mechanism such as CB06, but more detailed than the daytime, very near source mechanism originally developed by Olaguer (2012a,b; 2013). The intra-urban mechanism will include night-time chemical reactions involving nitrate radical. It will also include heterogeneous secondary formation of nitrous acid (HONO), an important HOx radical precursor, based on the parameterization of Sarwar et al. (2008) as modified by Fu et al. (2019). An accompanying semi-analytical chemical solver, with an explicitly derived chemical Jacobian matrix, will facilitate 4D variational data assimilation and inverse modeling based on the adjoint method.

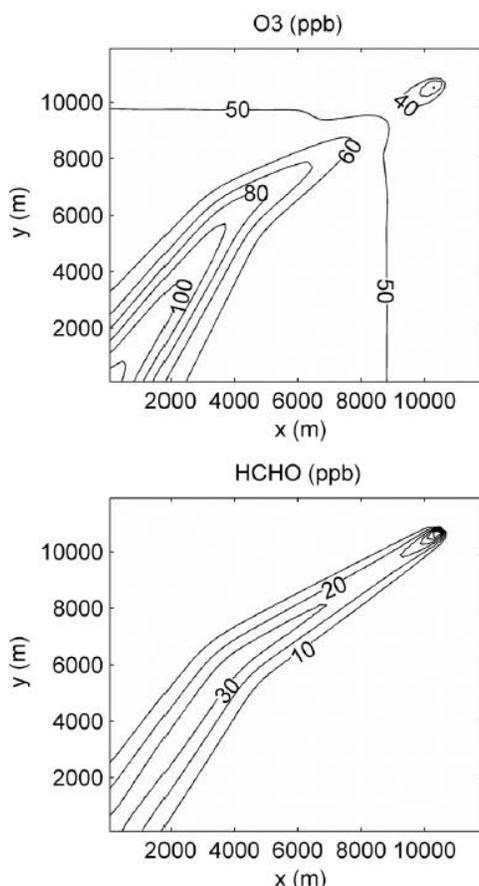


Figure 7. Concentration isopleths of ozone (upper figure) and formaldehyde (lower figure) generated by a large olefin flare event, as simulated by Olaguer (2012b).

The microscale air quality model will make use of building-sensitive wind fields from the Quick Industrial Complex (QUIC) model developed by Los Alamos National Laboratory, using Open Street Maps urban morphology data as input, or in select cases, more recent lidar-derived building data for key industrial facilities. Microscale modeling of ozone in the atmospheric boundary layer will be conducted using two 60 km x 60 km limited area fine mesh domains (see Figure 5). One microscale domain will cover the Detroit

metropolitan area in the southern part of the SEMI region (including Windsor), while the other will cover the northern SEMI region including Port Huron (along with Sarnia).

The Global Environmental Multi-scale (GEM) numerical model is ECCC’s operational meteorological forecasting model with a national domain and 2.5-km grid spacing (Milbrandt et al., 2016). The GEM-MACH model is a chemical transport model composed of dynamics, physics and atmospheric chemistry modules run on-line within the GEM model (Stroud et al., 2020). For this study, GEM-MACH will be run in nested mode, comprised of an outer national domain at 10-km grid-spacing, intermediate domain of 2.5-km grid spacing, and high-resolution domain of 1-km spacing encompassing the cities of Toledo, Windsor, Detroit, and Sarnia. In a recent development, the 2.5-km GEM model can now be used to create a meteorological analysis to initialize the higher resolution GEM-MACH cycles. The surface scheme in GEM-MACH is based on an advanced soil moisture and land surface temperature data assimilation system. Hourly lake water temperature is also assimilated into a lake model analysis. An urban canopy scheme, called the Town Energy Balance (TEB), is used to simulate the urban heat island effect (Ren et al., 2020). The impact of new, remotely sensed information on urban roughness will be compared to existing data for the Border region. Sensitivity studies with more resolved urban roughness information can assess the importance of uncertainties in the urban surface structure on urban meteorology.

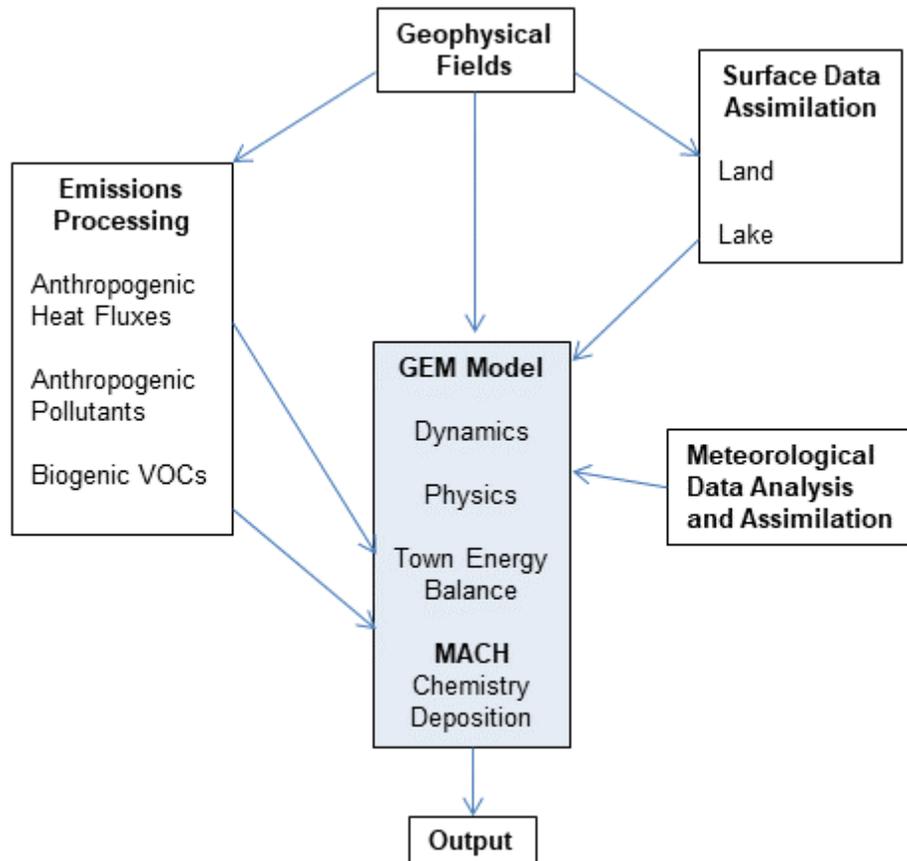


Figure 8. Schematic of processes in the GEM-MACH model.

Table 4. High-Resolution GEM-MACH model configuration and settings used for this study.

Numerical Model Option	Option Description
Grid Spacing	2.5-km × 2.5-km
Meteorology Data Assimilation	Ensemble variational (EnVAR) method
Cloud Microphysics	Milbrandt and Yau two-moment bulk
Longwave Radiation	Li-Barker correlated-k distribution
Boundary Layer Scheme	TKE with statistical representation of sub-grid clouds (MoistTKE)
Cloud Convection	Kain-Fritsch scheme, important for summertime convection
Land Surface Scheme	ISBA and Town Energy Balance
Surface Data Assimilation	CALDAS with ensemble Kalman filtering, hourly for temperature and moisture assimilation; 2-km NEMO model for lake with 10-km analysis
Gas-Phase Chemistry	ADOM-II mechanism
Gas-to-Particle Equilibrium	HETV (Heterogeneous Chemistry Vectorized)
Gaseous Deposition	Resistance model using Henry's Law and Oxidation Potential
Photolysis Rates	Look-up table and modulation based on cloud fraction
Physics Time Step	120 s
Chemistry Time Step	240 s

Figure 8 shows a schematic of the dynamics, physics, and chemistry processes represented in GEM-MACH-TEB. GEM-MACH-TEB includes a comprehensive chemistry process package that represents gas-phase chemistry, aqueous-phase chemistry, and particle microphysics (nucleation, condensation, coagulation, settling and deposition). Table 4 lists the key model settings for chemistry and physics.

For GEM-MACH simulations, the Canadian National Pollutant Release Inventory (NPRI) will be considered for point-source emissions, and the Canadian Air Pollutant Emission Inventory (APEI) will be used for area-source emissions. For the U.S., pollutant emissions will be obtained from the U.S. EPA National Emissions Inventory, with mobile emissions based on the MOVES traffic model.

The GEM-MACH study will analyze ozone exceedance periods in 2018 at the Windsor West site. The model will be validated with available air quality data in the Border region. A conceptual picture for ozone exceedance events will be created. Case study periods will be selected for future ozone attainment demonstration experiments. The sensitivity of the modelled 8-hr ozone maximum for the case study periods will be determined through a series of incremental emission perturbation simulations. The sensitivity of modelled ozone maxima to incremental NO_x emission reduction for a Border region domain will be determined. This will provide insight to the ozone production chemical regime (NO_x, VOC, or transitional). Incremental NO_x emission reductions by source sector can provide information on NO_x source apportionment in the Border domain. Similar sensitivity runs can be performed by VOC source sector with particular interest in the non-combustion sector as a whole (paints, glues, VCPs), as collectively it is larger than combustion sector in cities and it has the largest uncertainty (MacDonald et al., 2018). These incremental emission reduction simulations can be used to derive source apportionment by emission sector for select locations, and can be compared with receptor-modelling, such as PMF. Based on these sensitivity simulations and feasibility analyses, a series of emission reduction scenarios will be developed and applied to the case study periods to assess attainment.

The GEM model will be run at 250-m grid spacing for the Border region during the MOOSE study period to generate detailed wind fields and turbulence characteristics. These high-resolution model outputs can be used to constrain local-scale dispersion models. MECP uses these dispersion models for inverse-emission modelling of point sources using mobile laboratory measurements (see section 2.3.2). Emissions derived from both EGLE and MECP via inverse modeling will benefit both ECCC and EPA in their modeling and source apportionment assessments.

The team from Brown University, WSU, and SUNY-ESF will develop a state-of-the-art isotope-driven 0-D photochemical box model and a chemical transport model, constrained by MOOSE reactive nitrogen concentration and isotope field measurements, to improve our understanding of the chemical mechanisms of ozone formation. The isotope-enabled NEI will be implemented in CMAQ to quantify the impacts of the updated chemical mechanisms on urban to regional ozone air quality and secondary aerosol budgets.

REFERENCES

- Brook, J.R. et al., Exploring the nature of air quality over southwestern Ontario: main findings from the Border Air Quality and Meteorology Study, *Atmos. Chem. Phys.*, **13**, 10461–10482, 2013.
- Duren, R. et al., California's methane super-emitters, *Nature*, **575**, 180-184, 2019.
- Fu, X. et al., The significant contribution of HONO to secondary pollutants during a severe winter pollution event in southern China, *Atmos. Chem. Phys.*, **19**, 1–14, 2019.
- Herman, J. et al., NO₂ column amounts from ground-based Pandora and MFDOAS spectrometers using the direct-sun DOAS technique: Intercomparisons and application to OMI validation, *J. Geophys. Res. Atmos.*, **114**, <https://doi.org/10.1029/2009JD011848>.
- Judd, L.M. et al., Evaluating the impact of spatial resolution on tropospheric NO₂ column comparisons within urban areas using high-resolution airborne data, *Atmos. Meas. Tech.*, **12**, 6091–6111, 2019.
- Judd, L.M., Al-Saadi, J.A., Szykman, J.J., Valin, L.C., Janz, S.J., Kowalewski, M.G., Eskes, H.J., Veeffkind, J.P., Cede, A., Mueller, M., Gebetsberger, M., Swap, R., Pierce, R.B., Nowlan, C.R., Abad, G. G., Nehrir, A., and Williams, D. Evaluating Sentinel-5P TROPOMI tropospheric NO₂ column densities with airborne and Pandora spectrometers near New York City and Long Island Sound, *Atmos. Meas. Tech.*, **13**, 6113–6140, <https://doi.org/10.5194/amt-13-6113-2020>, 2020.
- Lake Michigan Air Directors Consortium (LADCO), 2017 Lake Michigan Ozone Study (LMOS): Preliminary finding report, Rosemont, IL, 2019.
- Makar, P. et al., Mass tracking for chemical analysis: the causes of ozone formation in southern Ontario during BAQS-Met 2007, *Atmos. Chem. Phys.*, **10**, 11151–11173, 2010.
- McDonald, B.C. et al., Volatile chemical products emerging as largest petrochemical source of urban organic emissions, *Science*, **359**, 760–764, 2018.
- McGill, M., Hlavka, D., Hart, W., Scott, V. S., Spinhirne, J., and Schmid, B. Cloud Physics Lidar: instrument description and initial measurement results, *Appl. Opt.*, **41**, 3725, <https://doi.org/10.1364/AO.41.003725>, 2002.
- McKain, K. et al., Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts, *PNAS*, **112**, 1941–1946, 2015.
- Milbrandt, J.A. et al., The pan-Canadian high resolution (2.5 km) deterministic prediction system, *Weather Forecast.*, **31**, 1791–1816, 2016.
- Nowlan, C.R. et al., Nitrogen dioxide observations from the Geostationary Trace gas and Aerosol Sensor Optimization (GeoTASO) airborne instrument: Retrieval algorithm and measurements during DISCOVER-AQ Texas 2013, *Atmos. Meas. Tech.*, **9**, 2647–2668, 2016.
- Nowlan, C.R., Liu, X., Janz, S.J., Kowalewski, M.G., Chance, K., Follette-Cook, M.B., Fried, A., González Abad, G., Herman, J.R., Judd, L.M., Kwon, H.-A., Loughner, C.P., Pickering, K.E., Richter, D., Spinei, E., Walega, J., Weibring, P., and Weinheimer, A.J. Nitrogen dioxide and formaldehyde measurements from

- the GEOstationary Coastal and Air Pollution Events (GEO-CAPE) Airborne Simulator over Houston, Texas, *Atmos. Meas. Tech.*, **11**, 5941–5964, <https://doi.org/10.5194/amt-11-5941-2018>, 2018.
- Olaguer, E.P., The potential near source ozone impacts of upstream oil and gas industry emissions, *J. Air and Waste Manage. Assoc.*, **62**, 966–977, 2012a.
- Olaguer, E.P., Near source air quality impacts of large olefin flares, *J. Air and Waste Manage. Assoc.*, **62**, 978–988, 2012b.
- Olaguer, E.P., Application of an adjoint neighborhood scale chemistry transport model to the attribution of primary formaldehyde at Lynchburg Ferry during TexAQS II, *J. Geophys. Res. Atmos.*, **118**, 4936–4946, 2013.
- Olaguer, E.P. et al., Attribution of primary formaldehyde and sulfur dioxide at Texas City during SHARP/ Formaldehyde and Olefins from Large Industrial Releases (FLAIR) using an adjoint chemistry transport model, *J. Geophys. Res. Atmos.*, **118**, 11317–11326, 2013.
- Olaguer, E.P. et al., Overview of the SHARP campaign: motivation, design, and major outcomes, *J. Geophys. Res. Atmos.*, **119**, 2597–2610, 2014.
- Phillips, N.G. et al., Mapping urban pipeline leaks: methane leaks across Boston, *Environ. Pollut.*, **173**, 1–4, 2013.
- Ren, S., Stroud, C.A., Bélair, S., Leroyer, S., Munoz-Alpizar, R., Moran, M.D., Zhang, J., Akingunola, A., Makar, P.A., Impact of urbanization on the predictions of urban meteorology and air pollutants over four major North American cities. *Atmosphere*, **11**, 969, 2020.
- Sarwar, G. et al., A comparison of CMAQ HONO predictions with observations from the northeast oxidant and particle study, *Atmos. Environ.*, **42**, 5760–5770, 2008.
- Seltzer, K.M., Pennington, E., Rao, V., Murphy, B.N., Strum, M., Isaacs, K.K., Pye, H.O.T., Reactive organic carbon emissions from volatile chemical products, *Atmos. Chem. Phys.*, **21**, 5079–5100, 2021.
- Stroud, C.A., Ren, S., Zhang, J., Moran, M., Akingunola, A., Makar, P., Munoz, R.A., Leroyer, S., Bélair, S., Sills, D., Brook, J. Chemical Analysis of Surface-Level Ozone Exceedances during the 2015 Pan American Games, *Atmosphere*, **11**, 572, 2020.
- Von Fischer, J.C. et al., Rapid, vehicle-based identification of location and magnitude of urban natural gas pipeline leaks, *Environ. Sci. Technol.*, **51**, 4091–4099, 2017.
- Zoogman, P., Liu, X., Suleiman, R. M., Pennington, W. F., Flittner, D. E., Al-Saadi, J. A., Hilton, B. B., Nicks, D. K., Newchurch, M. J., Carr, J. L., Janz, S. J., Andraschko, M. R., Arola, A., Baker, B. D., Canova, B. P., Chan Miller, C., Cohen, R. C., Davis, J. E., Dussault, M. E., Edwards, D. P., Fishman, J., Ghulam, A., González Abad, G., Grutter, M., Herman, J. R., Houck, J., Jacob, D. J., Joiner, J., Kerridge, B. J., Kim, J., Krotkov, N. A., Lamsal, L., Li, C., Lindfors, A., Martin, R. V., McElroy, C. T., McLinden, C., Natraj, V., Neil, D. O., Nowlan, C. R., O’Sullivan, E. J., Palmer, P. I., Pierce, R. B., Pippin, M. R., Saiz-Lopez, A., Spurr, R. J. D., Szykman, J. J., Torres, O., Veefkind, J. P., Veihelmann, B., Wang, H., Wang, J., and Chance, K. Tropospheric emissions: Monitoring of pollution (TEMPO), *J. Quant. Spectrosc. Radiat. Transf.*, **186**, 17–39, <https://doi.org/10.1016/j.jqsrt.2016.05.008>, 2017.